Optical Bistability in Colloidal Crystals

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We present a one dimensional model for the nonlinear response of a colloidal crystal to intense light illumination along a high symmetry direction. The strong coupling between light and the colloidal lattice, via the electric gradient force acting upon the particles, induces a novel large optical nonlinearity. We obtain bistable behavior when the incident frequency is inside the stopband of the periodic structure, with decreasing switching intensity as the frequency increases. The transmission characteristics and the magnitude of the switching threshold intensity are also in good agreement with a recent experiment.

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I. INTRODUCTION

Photonic band gap (PBG) materials [1] do not allow propagation of electromagnetic waves within a certain frequency range, thereby opening the possibility of studying new physics within the gap. In addition, many novel applications of these PBG crystals have been proposed, with operating frequencies ranging from microwaves to the optical regime [1,2]. Structures exhibiting full photonic band gaps in the microwave [3], millimeter [4] and submillimeter [5] regimes have already been fabricated, but scaling these structures down to the optical regime has remained a challenge. One way to construct PBG crystals in the optical regime is by growing polystyrene colloidal crystals [6–9], which have lattice spacing comparable to the wavelength of light. Such colloidal crystals do not exhibit a complete PBG, because the concentration and the index of refraction of the polystyrene spheres relative to water are not yet sufficiently high. However, they are very useful in studying PBG effects seen only in particular directions. In addition, they can be used in nonlinear optical studies. It is expected that the PBG effects can be strongly affected by nonlinear optical effects. In particular, it has been shown that intensity-dependent index of refraction can cause a shift in the locations of the band gap [10]. That is, if the index of refraction of either the suspended polystyrene spheres or water is intensity-dependent, then the width and the position of the stopband (or gap) will change under intense illumination. For example, a decrease of the index of refraction in water upon illumination will widen the gap, and, therefore, inhibits the propagation of the probe beam. Such an optical switching for light control is of great interest to the optics community.

In a recent experiment [7], optical switching and optical bistability were observed, when intense light was transmitted through a colloidal crystal. Simple switching was observed near the low-frequency end of the stopband, whereas bistability and multistability occurred at the center or near the high frequency end. The switching threshold was found to decrease as the incident frequency increased. These aspects are inconsistent with the response of a material with the conventional intensity-dependent nonlinearity [7]. The measured nonlinear coefficient $n^{'}$ inside the transmission band, $4\times10^{-10}cm^2/W$, is also several orders of magnitude larger than the electronic nonlinearity of both materials.

In this paper, we present a one dimensional model for the nonlinear response of a colloidal crystal to intense light incident along a high symmetry direction, based on the electrostriction mechanism [7]. Light is strongly scattered by the periodic arrangement of the colloidal particles inside the crystal, thus creating a spatially varying field. The polystyrene spheres, polarized by the electric field, will move in response to the electric gradient force. Such a structure change in turn will alter the propagation of light. The optical response is thus controlled by the the stationary configuration that results from the balance between the elastic and the electric gradient forces. We assume that the electric gradient forces are not strong enough to destroy the polystyrene spheres' crystalline structure and furthermore, that the changes in interparticle separation are small compared to the mean interparticle separation. Assuming the incident wave can be approximated as plane waves due to the large beam spot size, the structural change induced by light incident along a high symmetry direction will be primarily in the propagating direction. We neglect possible transverse effects and describe the three dimensional lattice by a one dimensional harmonic lattice model. Numerical calculations of the transmission characteristics, based on known physical properties of the colloidal crystal and a simplification to a layered structure, show good agreement with experiment. In particular, we find bistable behavior inside the stopband at intensities comparable to the observed switching threshold. The switching threshold is found to decrease as the incident frequency increases. We need to emphasize that such a nonlinearity necessarily depends on the exact stationary configuration of the lattice and therefore cannot be described with a simple effective intensity- and/or frequency-dependent dielectric constant.

This paper is organized as follows. In section II, we introduce our one-dimensional lattice model for the optical nonlinearity in colloidal crystal. In section III, we present results of calculations on the optical bistability and compare them with experiments. Conclusions and discussions are presented in section IV.

II. ONE DIMENSIONAL MODEL OF OPTICAL NONLINEARITY

In general, wave propagation in periodic structures is a complex phenomenon. Three dimensional scattering of light plays an important role in determining the nonlinear optical response of a colloidal crystal to incident light. However, simplification is possible if, a) the incident wave is plane-wave-like; b) the light is normally incident upon a high symmetry plane of the crystal lattice, and c) no transverse instability exits. Under these conditions, the structure can be viewed as a layered system. One dimensional modeling of the optical response is expected to be appropriate with correctly calculated physical parameters. The first condition ensures all spheres within one layer are equivalent, hence there should be no lateral lattice displacement, as required by symmetry. The second condition makes the layered structure more distinct since the distance between the layers is large. The third condition essentially requires that the structure is stable under illumination.

The colloidal crystal used in experiment [7] had a face-centered-cubic structure formed by polystyrene spheres (n_1=1.59) of approximately d_1=120 nm in diameter, at concentration f around 7%, dispersed in water (n_2=1.33). Light was normally incident upon the [111] plane which was parallel to the surfaces of the container. Due to the relatively large spot size, we approximate the incident wave as plane-waves. To a low intensity incident wave from the [111] direction, the fcc colloidal crystal acts essentially as a Bragg reflector (linear regime). The system is naturally simplified as a one dimensional bilayer structure consistent of alternating segments of polystyrene sphere layers (a mixture of polystyrene spheres and water with total thickness d_1) and pure water layers. With sphere concentration of 6.9%, the distance between the polystyrene sphere layers [11] is R_0=216 nm. The thickness of the water layer is then d_2=R_0-d_1=96 nm. The average index of refraction of the polystyrene layer is estimated [12] to be 1.36. Since the sample thickness is L=100 μm, the total number of bilayer units is N=L/R_0=463. The linear (zero intensity limit) transmission coefficient versus the wavelength in the [111] direction is shown in Fig. 1, calculated with the parameters mentioned above. Notice that there is excellent agreement [13] between our theoretical results of Fig. 1 and the experimental results of Fig. 2 in Ref. 7. This shows our model parameters describe very well the linear transmission of the colloidal crystal.

To illustrate that the nonlinear response of the colloidal crystal cannot be described by a one dimensional layered model with the conventional Kerr type nonlinearity (intensity-dependent dielectric constant), we show in Fig. 2 the nonlinear response of such a system, assuming that the effective index of refraction n_2 of the "water" has the form, $n_2 = n_2^o + n'|E|^2$. n_2^o=1.33 is the linear index of refraction of the medium and the nonlinear coefficient is taken to be the experimentally measured nonlinearity, $n' = 4 \times 10^{-10} cm^2/W$. Taking the propagation direction to be the z direction, we can solve the propagation of light governed by the following wave equation

$$\frac{d^2E}{dz^2} + \frac{n^2(z)\omega^2}{c^2}E = 0, \tag{1}$$

where n(z) is the index of refraction of our model which consists of alternating layers of linear and nonlinear medium, as specified above. Indeed, bistable behavior is obtained, as can be clearly seen in Fig. 2. The existence of such bistability phenomena in distributed feedback structures with intensity-dependent dielectric constants were predicted [14] theoretically and their properties have been investigated intensively [15]. Similar bistable behavior has been seen [16–18] in the discrete case of the electronic version of Eq. (2). However, the threshold intensity for the onset of the bistable behavior is in the order of 10 MW/cm^2, at least three orders of magnitude larger than the experimentally measured value of about 5 kW/cm^2. We see that a simple solution of Eq. (1) with an effective intensity-dependent nonlinearity indeed produces bistable behavior, but its predictions for the incident intensity threshold are unrealistically high. Clearly, a novel form of nonlinearity must be in action.

An interesting mechanism due to electrostriction was proposed [7] to be responsible for the nonlinear behavior in the colloidal crystal. In the absence of light illumination, the short-range screened electrostatic repulsive forces [19] between the spheres balances the weak long-range attractive force of Van der Waals type and produces an equilibrium configuration for the polystyrene spheres, with nearest neighbor separation $S_0 = \alpha/\sqrt{2}$ where α is the fcc lattice constant. When intense electromagnetic (EM) wave is incident upon the crystal, the dielectric spheres get polarized by the electric field and move in response to the gradient force [19] from the spatially varying field. This in turn alters the propagation of EM wave. Consequently, optical nonlinearity result. Ultimately, the optical response under a given illumination is controlled by the steady state lattice configuration which has to be determined by the balance of the elastic and electric gradient forces.

To determine whether the electrostriction mechanism is indeed responsible for the experimental observations, we have to solve simultaneously wave propagation equations and lattice dynamics incorporating both elastic and electric gradient forces. Here we propose a simple one

dimensional lattice model that can be solved straightforwardly but still contains the essential physics to account for the optical bistability observed in experiment. We have argued that under the experimental condition, a one dimensional layered model is appropriate to describe the transmission of wave along the propagation direction. The linear transmission property of this one dimensional lattice, consisting of alternating layers of polystyrene and water, has already been described (Fig. 1). To model the nonlinear response, we need knowledge of the lattice dynamics which is governed by the elastic and electric gradient forces.

We assume that for small fluctuations around the equilibrium configuration, the harmonic approximation is correct. We can then describe the motion of polystyrene spheres as if they were connected with each other with ideal springs. The force constant k of the springs can be roughly estimated by linearizing the screened electrostatic repulsive force [22],

$$F_{el} = \frac{(Ze)^2}{4\pi\epsilon R^2}\frac{1+\kappa R}{1+\kappa a}\exp[-\kappa(R-a)], \tag{2}$$

at the layer equilibrium position $R = R_0$. This leads to an expression $F_{el} = F_{harm} = k\Delta R$, where $\Delta R = R - R_0$ is the displacement from the equilibrium position. Z is the number of charges on the particle, κ is the inverse screening length, and a is the radius of the particle. Using $\epsilon = 1.33\epsilon_0$, $a = 60$ nm, $R_0 = 216$ nm, and assuming typical values [19] of Z=1000e$^-$ and $\kappa = 5\times10^7 m^{-1}$, we obtain k = 1.8×10^{-4}N/m. This corresponds to a bulk moduli B$\sim$ k/R_0 $\sim$1000 N/M^2, a reasonable value for colloidal crystals [19]. As we will see later, the electrostriction nonlinearity is inversely proportional to k. Only the order of the magnitude of k is relevant for our purpose. For definiteness, we take k = 1.8×10^{-4}N/m in the following calculations.

The gradient force on a sphere F_{gr} along the propagation direction z, can be calculated by taking the spatial derivative of it's polarization energy, i. e., $F_{gr} = -d(U_p)/dz$. A crude estimate of this force is

$$F_{gr} \simeq 4\pi n_1^2\epsilon_0\frac{m^2-1}{m^2+2}a^3\frac{1}{2}\frac{\Delta|E|^2}{2a} \tag{3}$$

where $m^2 = (n_1/n_2)^2$ is the dielectric contrast between polystyrene spheres and water, a is the radius of the spheres, and the factor $\frac{1}{2}$ comes from averaging over a time period. $\Delta|E|^2$ is the field intensity difference across a sphere's diameter. For our model's parameters this gives $F_{gr} \simeq C\Delta|E|^2$, $C = 2.2\times10^{-26}Nm^2/V^2$.

The optical response of the colloidal crystal is determined by the steady state configuration. In our one dimensional model, this is reflected as the steady state lattice configuration representing the configuration of the layers. Taking nearest neighbor interactions only and denoting by ΔR_n the change from the equilibrium separation of particles n and n+1, we have for the steady state that

$$F_{gr} = -F_{harm} = -k(\Delta R_n - \Delta R_{n-1}). \tag{4}$$

The gradient force F_{gr} on each polystyrene layer has to be calculated from the electric field distribution via Eq. (3) for the given lattice configuration $\{R_n\}$.

The transmission characteristics are obtained by solving Eqs. (1) and (4) self-consistently through iteration, for a given input. In actual calculations, however, this is done for a given output because the presence of bistable or multistable behavior. In a nonlinear one-dimensional model, each output corresponds to exactly one solution, while a given input may correspond to more than one output solutions (bistability). The input intensity can be reconstructed once the transmission coefficient is calculated after solving Eq. (1). We start with the equilibrium configuration in the absence of light in which all the layers is equally spaced with distance R_0. The wave field E(z) is then calculated from Eq. (1), with n(z) given by the exact one-dimensional lattice configuration $\{R_n\}$. n(z) equals to 1.36 if z is in the polystyrene layer and 1.33 otherwise. The gradient force and the elastic force is then calculated and R_n is increased or decreased depending the direction of the total force. The wave field is then recalculated and accordingly $\{R_n\}$ readjusted. This iteration procedure continues until the total force vanishes on each polystyrene layer. The final configuration will be the steady state configuration, and the corresponding field represents the actual optical response of the system. Twenty iterations are usually required before a steady state self-consistent configuration is achieved.

We point out that the present situation is analogous to the problem of an electron moving in a one-dimensional harmonic lattice with electron-lattice interactions. Such an analogy may help to understand the nonlinear optical response when the frequency is inside the stopband. We comment that neither in the polystyrene spheres nor in the water have we assumed any intrinsic nonlinearity. The nonlinear response of the colloidal crystal is entirely due to the coupling between the light and the lattice. In principle, such coupling exists in all materials. But the extreme softness of colloidal lattices relative to conventional crystals, reflected in the small value of the effective spring constant k, makes the observation of nonlinear effects possible in these materials.

III. OPTICAL BISTABILITY

As a first check of our model, we have numerically calculated the sign and the strength of the effective nonlinearity for a frequency (λ=514 nm) inside the transmission band. We found that the colloidal crystal linearly expands with the incident intensity of the EM wave, with a slope of about $1nm$ per $30kW/cm^2$. This corresponds to a relative linear expansion of the order $\Delta L/L \simeq 10^-5$, which is quite small as required by our harmonic assumption. The resulted phase shift in the transmitted wave

can be related to an effective positive nonlinear index of refraction by

$$\frac{\omega}{c}(n_2 - 1)\Delta L \simeq \frac{\omega}{c} n' |E_0|^2 L \qquad (5)$$

where $|E_0|$ is the incident intensity. For our system we estimate $n' \simeq 10^{-10} cm^2/\mathrm{W}$. This is in excellent agreement with the experimental value [7] of $n' \simeq 4 \times 10^{-10} cm^2/\mathrm{W}$, considering that the value of the force constant is only estimated with typical values of physical properties for colloidal crystals. We find that within the transmission band, the nonlinearity n' scales almost linearly with 1/k, but with no appreciable frequency dependence. The experimentally observed nonlinearity can be matched with the choice k$\simeq 4.4 \times 10^{-5} N/m$ and a corresponding bulk moduli B$\sim 250 N/m^2$. For definiteness we continue to use the initial estimated value of k. Changing value of k amounts to rescale the light intensities, since the actual contraction or expansion is controlled by the ratio of the elastic force and the gradient force, ie, only the ratio of the k and light intensity matters.

Multistability and switching threshold intensities are also correctly predicted within our model for frequencies inside the band gap. The local expansions and contractions of the lattice under illumination are the origin of the bistable behavior. Normally, transmission is forbidden in the gap of a periodic system. However, lattice distortion allows the existence of localized modes in the gap. Under appropriate conditions, the coupling of these localized modes with the radiation can produce resonant transmission. This is clearly seen in Fig. 3, where the local lattice expansion (a), the field intensity averaged in each sphere (b), and the intensity gradient (c), are shown as a function of the lattice plane, exactly at a transmission resonance for a frequency inside the stopband. Notice that there is a strong lattice deformation (solid curve in Fig. 3a) at the middle of the crystal, sustained by the strong field intensities (solid curve in Fig. 3b) and the intensity gradient. Similar behavior is seen for the case of the second transmission resonance (dotted lines in Fig. 3). This work clearly shows that there exists a strong light-lattice interaction, giving rise to lattice deformations which in turn produces localized solutions as "soliton-like" objects [20,21]. When these "soliton-like" objects appear symmetrically in the crystal, a transmission resonance is expected. Also, the longer the wavelength and the higher the multistability order are, the larger the maximum values of these deformations become. The bistable behavior originates from these field-distribution-specific structure changes. We point out that the total expansion of the lattice is still relatively small, generally in the order of 80 to 200 nm for each "soliton-like" object present in the structure.

The transmission characteristics are shown in Fig. 4, for four different wavelengths as were indicated in Fig. 1. We see that our model captures the most essential features of the nonlinear response of the colloidal crystal, as compared with the experimental results presented in Fig. 3 of Ref. 7. Notice that this model correctly predicts the magnitude of the switching threshold intensities, they are of the order of (20-40) kW/cm^2 and not of the order of 10^4 kW/cm^2 that the simple model with an intensity-dependent dielectric constant predicts (see Fig. 2) . The switching threshold intensities get smaller as we move from the long to the short wavelength side of the stopband, in agreement with experiment [7]. Bistability is observed when the lattice is distorted enough so to sustain a localized mode. This will happen if the local expansion is large enough to locally shift the effective gap to longer wavelengths [22]. Thus, the closer the incident frequency is to the small wavelength side of the gap, the smaller the lattice distortion needed to onset bistability, and thus the smaller the switching powers are.

Discrepancy with the experimental data is found for large incident intensities and in the low frequency side of the gap. Multistability was observed only in the high frequency side while for midgap frequencies the crystal is bistable and for low frequencies it is non bistable [7]. Also, at high intensities only instabilities were observed experimentally, in contrast to our model that predicts multistable behavior for all gap frequencies and all intensities. However, it is for the long wavelengths and the high intensities that the required lattice expansions get unrealistically large. The total lattice expansion versus the transmitted intensity are shown in Fig. 5, for the four wavelengths indicated in Fig. 1. Every local maximum in these curves corresponds to a transmission resonance. Since the crystal can not expand more than a certain maximum limit, an external pressure must be inserted into our model to limit its expansion. Numerical studies incorporating an external pressure show that while multistability is still predicted for all frequencies, the required local expansions and contractions, (with total expansion being constant and limited), and light intensities are much larger, making the starting assumption of a slightly perturbed harmonic lattice invalid. With large lattice distortions, approximation to a one dimensional structure also becomes questionable, and this may be the main reason for the discrepancy. The neglect of light absorption in water may also be a contributed factor to the discrepancy. Absorption reduces the light intensity nonuniformly, and thus may affect the nonlinear response.

IV. DISCUSSIONS AND CONCLUSIONS

We have shown that several essential features of the nonlinear response in colloidal crystals can be accounted for by a simple one dimensional model that incorporates the lattice distortions under intense light illumination. In this one dimensional model, the colloidal crystal is simplified as a one-dimensional layered system consisting of alternating layers of polystyrene spheres and water. The polystyrene layers represent high symmetry planes of the

colloidal crystal normal to the propagating direction and are modeled as elastic media deforming under the act of the gradient force from the electric field. Based on physical properties of the colloidal crystal, we are able to estimate the effective elastic spring constant. The wave equation of the electric field and the lattice dynamics of this one dimensional systems is then solved simultaneously to obtain the steady state response. We are able to obtain the correct order of magnitude of the effective nonlinearity within the transmission band and the switching intensity for optical bistability within the stopband. The trend that this switching intensity decreases as the frequency increases across the stopband is also reproduced. Although it seems surprising that a simple one-dimensional model works when three-dimensional scattering of light plays an important rule, detail considerations suggest this simplification should be appropriate under the experimental condition.

In conclusion, we have established with a simple one dimensional model that the light-lattice coupling via electric gradient force underlies the large optical nonlinearity observed recently in colloidal crystals. Such a coupling alone can produce bistability and multistability with switching threshold intensities and transmission characteristics in good agreement with experiment. Given the unique large nonlinear response, colloidal crystals may prove to be very useful for future studies of nonlinear effects in PBG materials in the optical regime.

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[11] For an fcc lattice of spheres of filling ratio f, with diameter d_1 and lattice constant α, the following relation is obeyed, $f=4\frac{(4\pi/3)(d_1/2)^3}{\alpha^3}$. The distance between the [111] planes in an fcc lattice is $R_0 = \alpha/\sqrt{3}$, and since d_1=120 nm, one easily gets that $R_0=88.64/f^{1/3}$ nm. So for f=6.9%, $R_0 \simeq$216 nm.

[12] The polystyrene layer is a mixture of polystyrene spheres with diameter d_1 and water. To better approximate the experimental system, it is appropriate to use an average index of refraction $\bar{n}_1$. The volume fraction f_1 of the spheres within the polystyrene layer is $f_1=R_0f/d_1$ = 12.4%. From $\bar{n}_1 = n_1 f_1 + n_2(1 - f_1)$, we determine $\bar{n}_1 \simeq$1.36.

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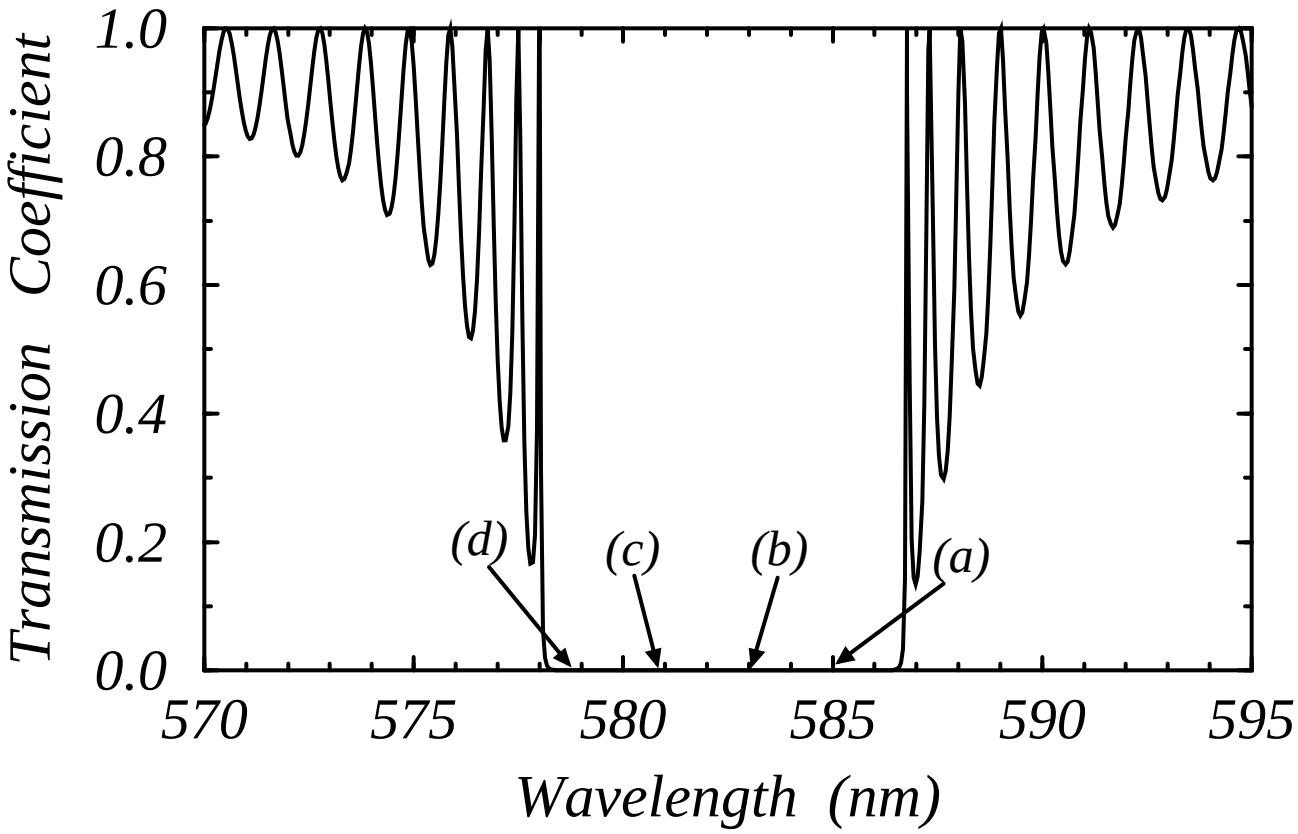
FIG. 1. Transmission coefficient versus the wavelength in the [111] direction of a colloidal crystal. The width of the polystyrene sphere layer is d_1=120 nm, with an effective [12] index of refraction $\bar{n}_1 \simeq$1.36, while the width of the water layer is d_2=96 nm with $n_2=n_2^o$=1.33. The number of bilayer units is N=463, which gives a sample thickness L=100μm. All the parameters used are in agreement with experiment. The arrows indicate the wavelengths of the incident light at which the transmission plots in Fig. 4 were obtained.

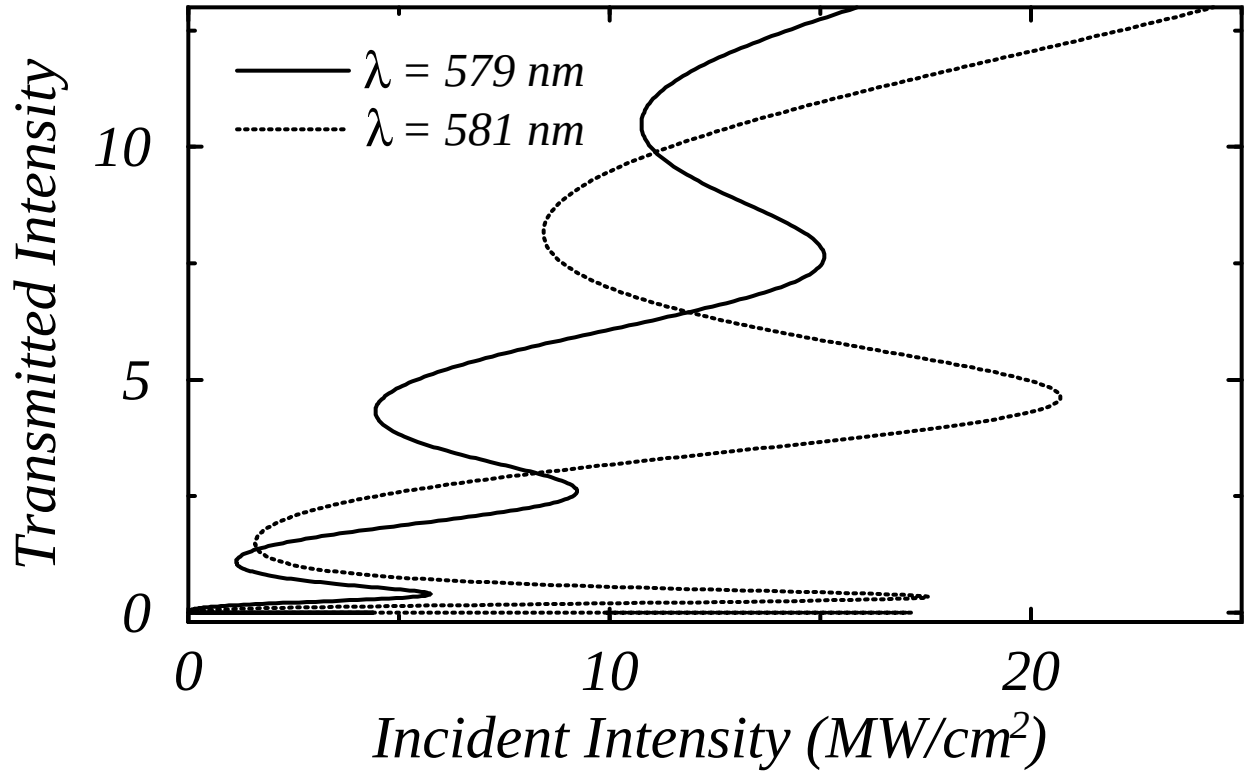
FIG. 2. Transmitted intensity versus incident intensity for a simple nonlinear bilayer system with $n' = 4 \times 10^{-10}\text{cm}^2/\text{W}$ (in water) for two values of the incident wavelength. This model gives unrealistically high values for the threshold incident intensities.

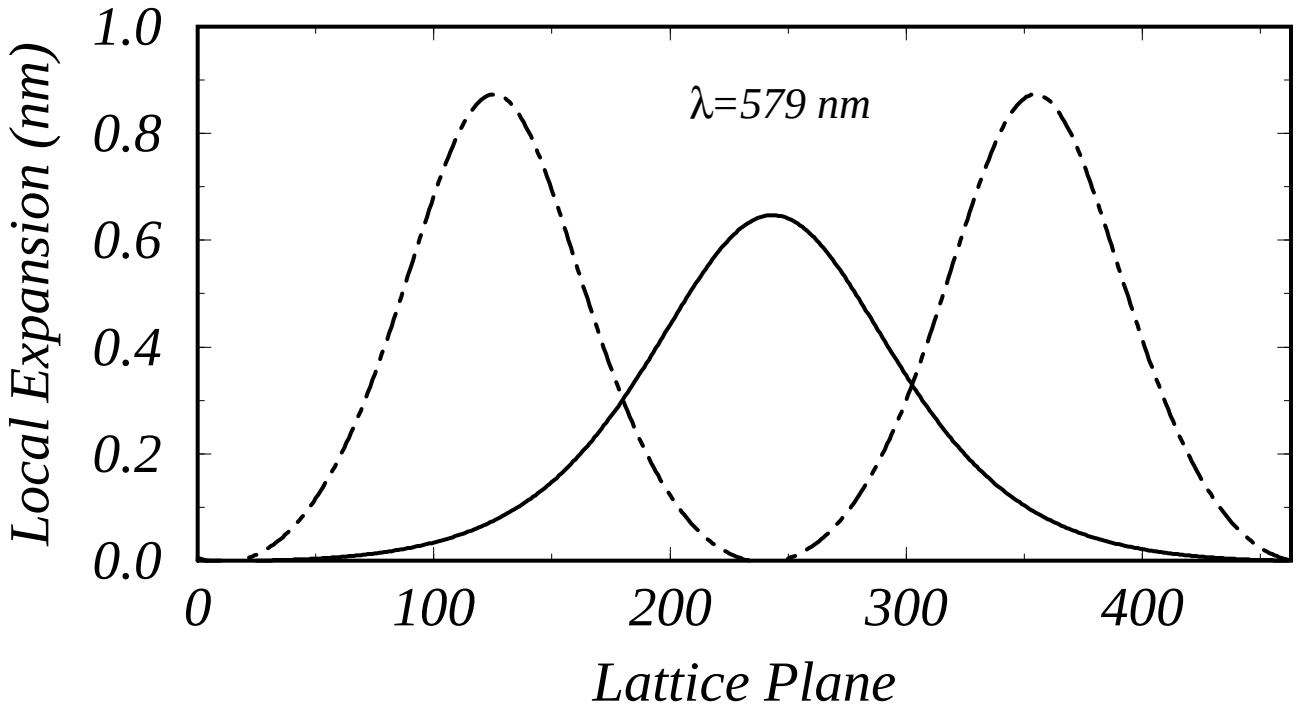
FIG. 3. Local lattice expansion (a), Field intensity averaged in each sphere (b), and Field intensity gradient (c), as a function of the lattice plane for λ=579 nm. Solid and dashed curves correspond to the first and second transmission resonances.

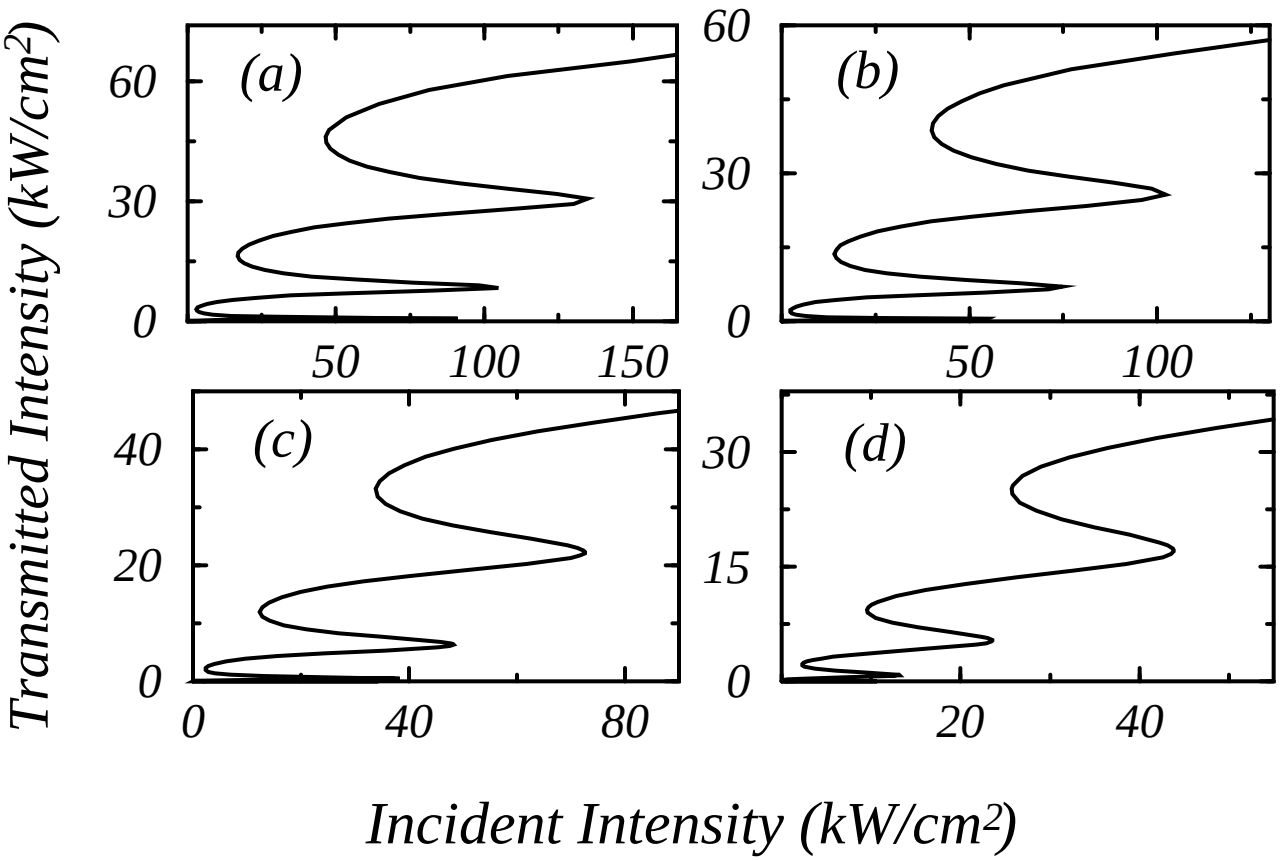
FIG. 4. Transmitted intensity versus incident intensity for four different wavelengths as were indicated in Fig. 1.

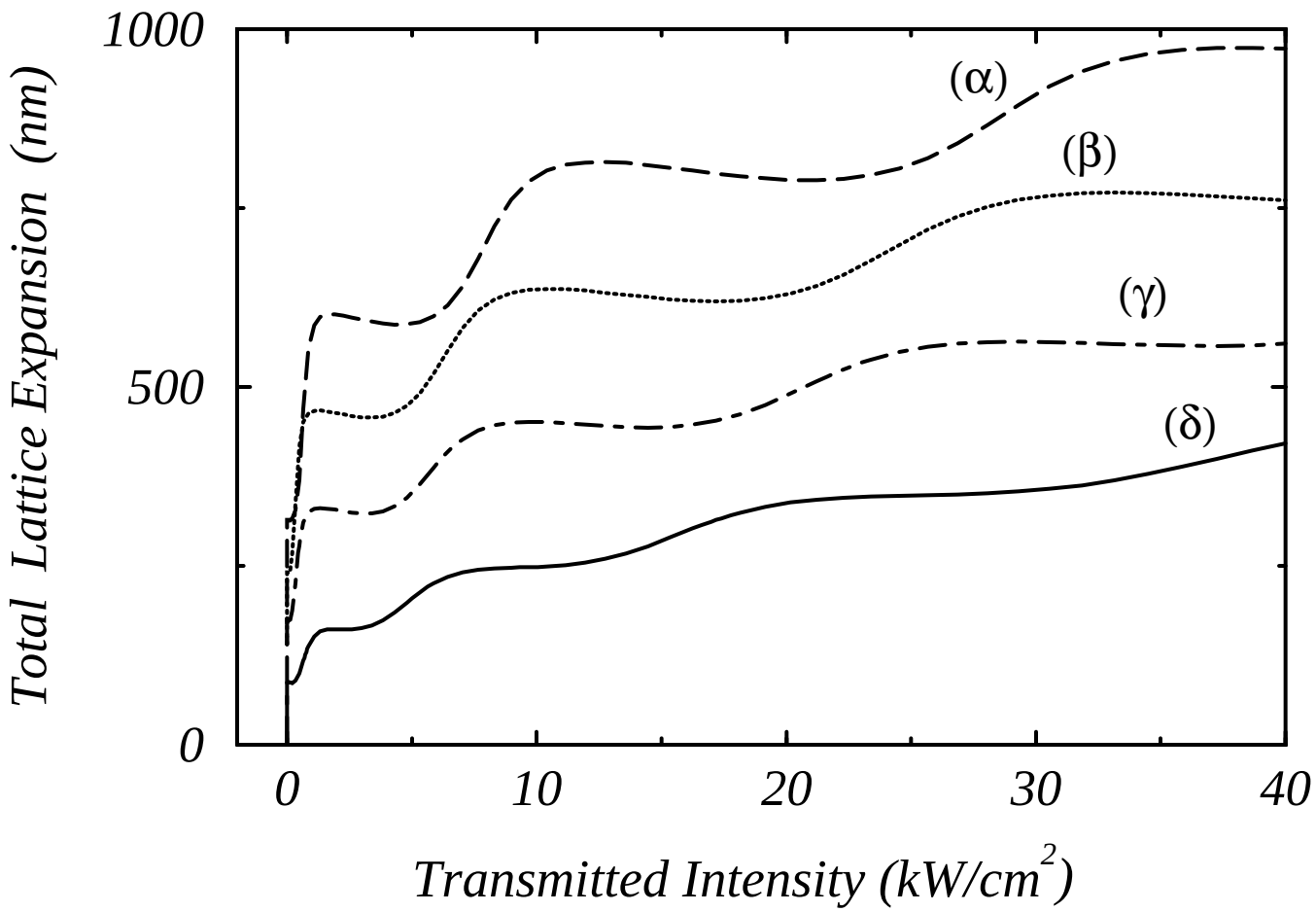
FIG. 5. Total lattice expansion versus transmitted intensity for the four wavelengths indicated in Fig. 1. No external pressure is assumed.


Transmission Coefficient
1.0
0.8
0.6
0.4
0.2
0.0
570
575
580
585
590
595
Wavelength (nm)
(d)
(c)
(b)
(a)


λ = 579 nm
λ = 581 nm
Transmitted Intensity
0
5
10
0
10
20
Incident Intensity (MW/cm2)


1.0
0.8
0.6
0.4
0.2
0.0
Local Expansion (nm)
λ=579 nm
0
100
200
300
400
Lattice Plane


Transmitted Intensity (kW/cm²)
(a)
(b)
(c)
(d)
60
30
0
50
100
150
60
30
0
50
100
40
20
0
0
40
80
30
15
0
20
40
Incident Intensity (kW/cm²)


1000
500
0
Total Lattice Expansion (nm)
(α)
(β)
(γ)
(δ)
0
10
20
30
40
Transmitted Intensity (kW/cm2)